Coal as a chemical feedstock

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Please cite as: CHEMIK 2015, 69, 12, 805–814

Back to the past

When we try to approach the problem briefly: "coal for chemistry" we immediately recall known relation – both hard coal and brown coal are mainly power carriers, and to much smaller extents feedstock to be used in chemistry. Such an approach makes it difficult to place coking industry in one branch of industry. Formally it is included in chemical industry, despite the fact that products of classic coking only to a small extent can be used as a feedstock for this industry, while the main product of the process – coke – is used in metallurgy.

Therefore, it might be worth it to return to old ideas presented on the mentioned figures that very clearly show coal as a chemical feedstock. An example of one them, from 1951, is presented in Figure 1 [1].

It is worth noting that proposition coal as a chemical feedstock (Fig. 1) is implemented through coking process, thus it applies only to coal mixtures of appropriate coke-forming properties, and to all types of coal, including lignite. Figure 1 presents typical product of classical coking process (in parentheses average share in relation to dry charge): coke (75–76%), coal tar (3.5–4.5%), benzole (approx. 1%), coke-oven gas (14-18%) and ammonia (0.2-0.4%). Figure 1 does not take into account sulphur compounds, mainly hydrogen sulphide, which expressed as elemental sulphur is approx. 0.2%. Constant evolution of coking technology resulting from the development of knowledge and resource demand causes that in modern coking plants ammonia and hydrogen sulphide contained in coke-oven gas are decomposed to elemental components. Ammonium in catalytic ammonia decomposition complex is decomposed to (inert) nitrogen, while hydrogen sulphide in Claus disproportionation process in the presence of SO₂ is oxidised to elemental sulphur of very high quality.





Fig. 1. The views on coal as a chemical feedstock acc. to [1] (the original version)

The wider scheme for processing of high-temperature tar was presented by Collin *et al.* [2, 3], which in Polish version is presented in Figure 2 [4].

Corresponding author: Professor Andrzej MIANOWSKI – Ph.D., D.Sc., (Eng.), e-mail: andrzej.mianowski@polsl.pl The analysis of idea presented in Figure 1 brings the conclusion that rejection of possibility of obtaining feedstock for chemistry through coking process leaves only one route: "products of largescale organic synthesis".

This paper does not attempt to review all known technologies of chemical coal processing in an encyclopaedic manner, but rather tries to answer the question: why in Poland coal is virtually a feedstock only for industrial and municipal power engineering? Maybe only is too big word – after all currently there is a production of products from distillation separation of benzole to BTX fraction in quantity not larger than 100,000 Mg/year at the plant "Petrochemia Blachownia".



Fig. 2. Comprehensive processing of typical coal tar acc. [2 - 4]

Update on coal-based products from the coking process

According to forecasts, in 2020 global coking industry will produce 925.3 mln Mg of coke, using 93% of total production capacity [5], while global coke consumption in 2013 was equal to 690 mln Mg [6]. This means that many countries still produce high-temperature tar. It is a feedstock for further processing by means of Continuous Distillation of Coal Tar or a final product intended for export. Starting from 2008, i.e. since the closing of the last Continuous Tar Distillation installation at Chemical Plant Blachownia, Polish coal tar is in total exported. Processing of liquid coal-based products in Poland is limited only to benzole refining and distillation. This way, for the purposes of the coking industry, more specifically for benzole absorption from coke-oven gas we need to import scrubbing oil, which is produced abroad from coal tar by means of distillation. We can assume there is a potential technological possibility of execution of some routes to products presented in Figures 1 and 2, as well as there are possible modifications in the processing of both coal tar, as well as grading benzole or its fraction by means of hydrofining. There are opinions that due to the nature of tar as a problematic feedstock for the processing industry, it would be most beneficial to subject it to hydrocracking

[7], destruction under supercritical thermal and pressure conditions [4] or use it as an admixture to heave petrochemical fractions, e.g. soft asphalt and then process them together [8, 9]. However, these concepts are verified only at research level.

Current considerations on reactivation of coal tar processing in Poland involve project of processing capacity of 200,000 Mg of tar/year, which with reserve corresponds to the level of tar production in coking plants owned by JSW KOKS S.A. (Fig. 3) [10].



Fig. 3. The block diagram of coal tar processing plant in JSW KOKS S.A. acc. [10]

Comparison of ideas (presented in Fig. 2 and 3) one can see the modification foreseeing that final products will be phenols and distilled naphthalene, with simultaneous covering of scrubbing oil demand for coking plant needs. It is also expected that solvent naphtha will be combined with benzole. Instead of "classic" coal-tar pitch, there will be rather large amount of distillation residues – 150,000 Mg/year (75%), which will be used by delayed coking plant. Currently, implementation of the delayed coking process due to technological and economical reasons is beyond the capacity of the Polish industry. Such is also a tone of comments on this issue presented in the paper [10].

It shall be noted that in comparison (Fig. 2 and 3) of distillation residue amount according to the concept [10], there is an increased quantity of anthracene oil, which for many years has been valuable component of oils for carbon black products at one time produced in Poland by channel method. It is worth to quote opinion of Z. Stompel [11] that long-term perspective of coal tar as chemical feedstock are justified if it will serve as a source of electrode tar pitch for aluminium and steel processing industry, naphthalene for production of plasticisers, gypsum and concrete and tar oil for carbon black production. Project [10] completely fulfils the position [11], while for production of carbon black further separation processes of anthracene oil by means of distilling off during the cooking process will be necessary.

A specific coking process product is coke oven gas often referred to as COG, which contains up to (in % v/v): 60% of hydrogen, 28% methane and 10% of carbon oxide [12]. For burning bank of coke ovens, coke oven gas after deeper purification is used, in quantities below 50%. The rest, called surplus coke oven gas is used in different power production options as well as fuel gas and reducing gas in iron metallurgy (DRI). Ready solutions and projects of separation of surplus hydrogen by means of pressure swing absorption (PSA) or its direct use for production of methanol [12, 15] dimethyl ether (DME) [15] are partially implemented, while others are still waiting for implementation.

Global trends of revolutionary changes in coking technology lead to dual-product coking plants. In such case, only products are coke and power, in this way eliminating typical coal-based products, i.e. tar, benzole and coke-oven gas [16]. Therefore, development of this idea in perspective will stop coking plants from being source of feedstock for chemical industry.

Coal liquefication and gasification

Despite historical background, it must be remembered that in 1940s Germany had plants for coal liquefication with gaseous

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hydrogen using Bergius method in 18 objects and Fischer-Tropsch (F-T) gasification method was used by 9 plants, with total liquid fuel production amounting to 4 mln Mg [17]. Certainly, F-T technologies are more attractive.

Direct hydrogenation of coal tar paste with gaseous hydrogen within Government Programme PR-1 named: "Comprehensive Coal Processing" conducted in the years 1976–1986 under the of auspices of prof. Jerzy Szuba found its final in the construction of pilot facility in Tychy-Wyry of processing capacity 1.2 Mg/day and significant development of chemistry and chemical coal technology [18]. However, nowadays in the face of increasing environmental requirements and growth of automotive industry and production of high-grade petrochemical motor fuel, carbon is not attractive feedstock for liquefication processes. Leaving aside the problem of hydrogen availability, the main issue concerns applicable standards for sulphur content (below 10 ppm), standardized content of aromatics and other physical and chemical properties [17]. Coal oil obtained in coal liquefication process, even after thorough hydrogenation, does not correspond in any way to group composition of typical petrochemical products.

We also have to mention methanation process, i.e. hydrogenation of coal to methane (C + $2H_2 = CH_4$). In Polish conditions it is unjustified, as in our technologies we use methane from natural gas for steam conversion in the presence of oxygen (POX process) to produce syngas, i.e. gaseous mixture of $H_2 + CO$.

New generation of coal processing technologies are referred to as CCT, i.e. Clean Coal Technologies. Coal gasification in the modern version of the POX involves combination of endothermic process of steam gasification with exothermic reaction of partial combustion following the equation: $C + H_2O + \frac{1}{2}O_2 = CO_2 + H_2$, of resultant, exothermic standard enthalpy $\Delta H^o_{298} = -165.1$ kJ/mol.

Main gasification production is syngas, which as shown in Figure 4 serves as a feedstock for further processing. Depending on the chosen direction of processing, it is necessary to establish appropriate hydrogen:carbon monoxide ratio with providing necessary purity of these substances.

If we consider methanol production, one can contemplate its application in chemical industry, even in abandoned process of Mobil Oil. As shown in Figure 4, this is represented by MTG (Methanol to Gasoline) technology, however, this process involves formation of large quantity of water (at least 56%) next to produced light petrol in amount below 40% [20]. Unsatisfactory cost-effectiveness of this idea in Polish conditions, can be summed up shortly: it is a waste to use hydrogen to produce useless water just to produce some petrol component.



Fig. 4. Possibilities of syngas application acc. [19] (variant analysed in [17])

MTP (Methanol to Propylene), which is a specific case of MTO (Methanol to Olefins) technology, may be considered attractive. Propylene after catalytic hydroformylation with syngas gives OXO alcohols. Among the products mentioned in the article [21], certainly the most important one is also DME (dimethyl ether).

Figure 4 shows other very attractive technological possibilities that we may consider unprofitable today in Polish conditions. We have to mention all F-T technologies, which definitely differ in catalyst quality (iron, cobalt, nickel catalysts, also with ruthenium) and temperature conditions, but still used in very matured form in the Republic of South Africa. Sasol concern, discussed in a fragment of paper [22] with focus on its current situation is an example of commercial solution of coal gasification process.

Meanwhile, in Poland, a fuel deficit (mainly of petrol and diesel fuel) that could be solved by implementation of F-T synthesis, was resolved by programme of Grupa Lotos, called 10+, which means increase in import and crude oil processing capacity to the level of 10.5 mln Mg/year. Within this program, ROSE (Residuum Oil Supercritical Extraction) facility was executed, where by means of close-to-supercritical extraction crude oil distillation residues are used to produce De-Asphalted Oil (DAO), which is ten used as a feedstock to Mild Hydrocracking (MHC) installation where (mainly) diesel oil is produced [4].

CTT euphoria and crisis

With Polish accession to the EU, intensive analyses of CCT has begun, including mainly "zero-emission" application of coal and development of coal-based chemistry. Completely new elements in this field apply to solutions for power industry, investing in IGCC (Integrated Gasification Combine Cycle) installations composed of gas and steam turbine, which in the case of chemical development are related through syngas with F-T technologies, methanol production or hydrogen separation. In each case, it was necessary to take into account Carbon Capture & Storage (CCS) facility. In the first, power-oriented case, heat and power generating plant Lublin-Wrotków showed great interest, but did not achieve the intended objective. One can easily summarize power production projects involving CTT by quoting statement issued in 2013 by PGE GiEK S.A., which informs that despite the large financial commitment it withdraws from the project CCS ready, as there is no:

- a) complete financing structure,
- b) implementation of CCS Directive.
- This way power industry said clearly farewell to CCT ideas.

Similar end met ideas of reducing deficit of liquid fuels using F-T technology, despite considerable scientific advances of the team of late Professor Wojciech Jóźwiak, who died in 2012 (Institute of General and Ecological Chemistry of Lodz University of Technology). Thorough studies regarding development of in-house catalysts for these syntheses, mainly bi-oxide catalysts were presented in only monograph available in Polish [23]. However, completely different reasons brought end to the attractiveness of F-T syntheses. In original Sasol solution, coal feedstock to final commercial product ratio, i.e. petrol and diesel fuel under their conditions is equal to: 3.3 : I and 3.4 : I, respectively, while acc. to [24] for low-temperature F-T synthesis it is much higher -5.7: I. This means that if we consider only production of diesel fuel, production yield is 3.4%, while if we consider also intermediate (which is always produced in much larger quantities) for production of diesel fuel, it is 14.2%. In total balance considering need of hydrocracking of solid paraffins (waxes) from low-temperature F-T synthesis for variant from feedstock to gas station, a final ratio can amount up to 10 : I [25]. This means that for expected quantity of 3 mln Mg/year of fuels (planned location in Oświęcim) requires inconceivable quantity of coal of constant quality. As mentioned above, liquid fuel deficit is resolved by 10+ programme by Grupa Lotos.

As a result of conflicts for performance of the contract for natural gas supply from Gazprom, former Zakłady Azotowe Puławy initiated advanced actions within CTT for production of hydrogen, and Zakłady Azotowe Kędzierzyn started to work on Poligeneracja Kędzierzyn. In the latter case, the highest development stage was achieved, with assumptions and block diagram of this concept is presented in Figure 5. Assumptions include that main products will be methanol in quantity over 500,000 Mg/year (thus completely eliminating need for import) and produced electricity. A novel and non-typical component in this

concept is use of hydrogen turbine at such a scale of gasification process, while currently on the market only Mitsubishi turbines are available with capacities lower than assumed ones. The further development of the program was stopped mainly due to the lack of progress in the field of CCT (due to the reasons similar as ones presented above).



Fig. 5. Assumption of "Poligeneracja Kędzierzyn" project (source: E. Sutor [26], with the permission of the Author)

Therefore, an answer which production variant is more promising, production of methanol or hydrogen lies in the field of the CCTs for chemical applications. Detailed and basic process data is presented in the feasibility study [24]. It seems intuitively that we shall choose methanol, as it is hard to foresee in what manner hydrogen will be used. Another variant also seems very interesting – production of fertilizers through synthesis of ammonia and, further, urea. However, this tendency will be justified only for supply of cheaper coal-based syngas than natural gas-based syngas.

Many other opinions also puts methanol from coal in the first place of cost-effectiveness in comparison with power production and production of motor fuels by means of F-T syntheses and as comparable to production of hydrogen.

Coal as a feedstock for chemistry development

All CCT concepts, after rather detailed preliminary analyses, have been abandoned. Certainly, moments of Gazprom threats and changing natural gas prices (obviously rising) drive ideas for production of hydrogen or methanol (which can be considered "liquid hydrogen") for chemical applications. If in the mid-2012 gas price in Poland was \$526 for 1,000 m³ (at that time the lowest was in the UK \$313), today the price dropped to \$225/1,000 m³, which can be related to diversification of gas supply, from EU and future supplies at LNG terminal in Świnoujście.

In 2010, works within strategic programme guided by the AGH University of Science and Technology were initiated within Science and Research Consortium "Coal Gasification" named: "Development of coal gasification technology for highly efficient production of fuels and electricity under the strategic research and development programme" which was completed in September 2015. In terms of "surface" gasification, the research resulted in the Institute for Chemical Processing of Coal developing an original process of pressure coal gasification in reactor with cylindrical fluidised-bed, with CO₂ as a gasification agent [27, 28].

Block diagram of process concept is presented in Figure 6 [27], which presents alternative solutions both for chemical applications (involving methanol production) and power industry – power production at the IGCC plant. Characteristic feature is combination of gasification island in POX version with CO_2 additional stream, partial gasification (with pyrolysis) and oxygen combustion of obtained devolatilized char. Therefore, it is a Polish original version of gasification process, which may be called POX_bis.



Fig. 6. The concept of coal gasification with CO₂ in a fluidized bed reactor to electricity A and methanol B production acc. [27]

For methanol production, gas composition after treatment shall have the following ratio $(H_2 - CO_2)/(CO + CO_2) \approx 2$, as addition of CO₂ promotes synthesis at reduced pressure. Thus, composition of syngas (presented in Fig. 5) including CO₂ is justified.

It must be noted that technological analyses, i.e. process designs and preliminary feasibility studies were prepared for many variants, involving hard coal and lignite, various final products (production of electricity/heat or methanol) and scale of production (demo installation 115,000 Mg methanol/year or industrial installation 520,000 Mg methanol/year).

Grupa AZOTY returns to a polygeneration, i.e. its new variant for ZAK SA, but in significantly modified form (in comparison to one presented in Fig. 5). On one hand, in-house solutions were created, on the other, it is planned to purchase licensed technologies.

Certainly technology acc. to [26, 27] is thoroughly worked in the design version. According to the criteria called readiness level, the Author assesses it to be at level 6 of 9 level possible and a big steps towards commercialization have to be certainly related with possibility of construction of demo installation, i.e. to achieve level 7.

Future perspectives for coal as a chemical feedstock in Poland

To be more precise, subheading shall state "coal as a abandoned chemical feedstock". In view of today's reality in the nearest time horizon, all the projects will not leave concept stage, still staying far from even complete feasibility studies. I repeat for a third time, that in post-War Poland the following motto is still up to date: "Poland lies on coal and lies with coal" [29, 30]. We must add also other professor's opinion that we shall not disregard our valuable raw material, because if abandon construction of nuclear power plants, we are left only with coal as a source of energy and chemical feedstock [31]. According to the report of the Industrial Development Agency (ARP SA) branch in Katowice, dated 4th September 2015, net financial result of the mining industry for the first half of 2015 was - 1,445 mln PLN. Mining industry crisis affects also JSW SA, as a result project of return to coal tar processing (Fig. 3) is postponed ad Kalendas Graecas. Only dramatic situation involving lack of export contracts for tar, can make owners of coking plants take equally dramatic actions, to a full extent, including also implementation of delayed coking (technology not used in Poland). In a critical situation, it will be necessary to either close the coking plants, or unofficially burn the tar on industrial scale (iron blast furnace, oil compositions for cement plants).

Despite the crisis, it seems realizable to have chemical processing of not more than from I to 2 mln Mg/year of hard coal or, preferably, lignite (it contains up to 50% of water) for gasification process in the POX_bis version According to Z. Kasztelewicz [32], exploited lignite deposits in Bełchatów, Turów, Konin and Adamów will be exhausted by 2040. Certainly, new lignite mines will be established in the area of Legnica-Ścinawa, Ruja, Gubin-Brody-Mosty and maybe in other locations on identified deposits in Greater Poland. Location of mines dictates also localisation of chemical plant, which enables for prioritising actions of KGHM Polska Miedź.

One may think that this works presents CTTs in an excessively simplified manner, particularly possibility of using syngas, methanol and further process sequences, mainly in the field of C₁ chemistry. However, this is done on purpose, in order to present broader background of the issue in Poland: hard coal, or maybe lignite as feedstock for large-scale organic synthesis (Fig. 1). Such detailed process dendrograms one can find in the papers [19, 21, 33 – 35]. The list of references also indicates this are Polish positions. In fact, each of them was created based on a review of global technologies, which is best exemplified by two last monographs printed within the project "Coal Gasification" [36, 37]. They form a very detailed compendium of knowledge in the field of coal preparation and gasification. These two book are a great introduction for all to all the global technologies.

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Conclusions

- At the end of 2015, coal as a chemical feedstock has its small share in Polish chemical industry in products of benzole distillation in the quantity of up to 100,000 Mg/year. At most, we return to the idea of production of methanol or hydrogen.
- More and more restrictive environmental policy of the EU, mainly in terms of CO₂ emissions will make us reconsider development of power industry based on CCTs, which will become an important point for development of licensed technologies in Grupa AZOTY on industrial scale, or following POX_bis technology as demo installation.

The review was prepared under the project "Coal gasification" funded by National Centre for Research and Development (NCBiR) (agreement No. SP/E/3/7708/10)

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Nowy skład Komitetu IChiP PAN w kadencji 2015-2018

Na posiedzeniu Komitetu Inżynierii Chemicznej i Procesowej PAN kadencji 2011–2015, które odbyło się 20 listopada 2015 r. na Wydziale Inżynierii Chemicznej i Procesowej Politechniki Warszawskiej ustalono, że dokonany zostanie wybór 24 członków tego Komitetu na kadencję 2015–2018. Do Komisji Wyborczej wpłynęło 112 zamkniętych kopert z wypełnionymi kartami do głosowania. Pośród nowo wybranych 24 członków Komitetu IChiP PAN na kadencję 2015–2018 jest Pani prof. dr hab. inż. Bożenna Kawalec-Pietrenko z Politechniki Gdańskiej, prezes Zarządu Oddziału w Gdańsku i Wiceprezes ZG SITPChem. Gratulujemy! (*abc*)

(inf. redakcji, 30 listopada 2015)

RYNEK

TAURON: Sukces w pozyskiwaniu finansowania

TAURON pozyskał nowe źródło finansowania w postaci programu emisji obligacji na łączną kwotę 6,27 mld PLN z gwarantowanym objęciem przez banki. Program będzie obowiązywał przez pięć

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Since 2006 he is a professor at the Institute for Chemical Processing of Coal (IChPW) in Zabrze. He specialises in coal processing technologies and utilisation of solid waste, as well as – thermal analysis in technological and industrial aspect. He has authored or co-authored 140 papers, 26 patents (including I utility pattern) obtained, with several implemented. He has also co-authored 8 chapters in books, scripts and over 100 presentations. He served as a supervisor for 10 doctors of technical sciences, three of whom have received distinction. Permanent Reviewer of the Journal of Thermal Analysis and Calorimetry". Since 2004 – he serves as a member of Editorial Committee of journal "Karbo" and a member of Programme Board of journals "Przemyst Chemiczny"; since 2012 of journal "CHEMIK"; since 2013 member of Programme Board "УГЛЕХИМИЧЕСКИЙ ЖУРНАЛ" (Ukraine)..

He was a member, or still is a member, of the following organisations: The Middle Pomeranian Scientific Society of the environment protection (until 2010), the Polish Society of Calorimetry and Thermal Analysis (until 2009), Polish Carbon Society (since 2010).

lat z możliwością dwukrotnego przedłużenia o rok za zgodą stron uczestniczących w transakcji. Jego uczestnikami są PKO Bank Polski, ING Bank Śląski, Bank Zachodni WBK, Bank Handlowy w Warszawie, Bank of Tokyo-Mitsubishi UFJ (Holland), Bank of Tokyo-Mitsubishi UFJ (Polska), CaixaBank Oddział w Polsce, Industrial and Commercial Bank of China (Europe) Oddział w Polsce. Funkcję agenta emisji pełni Bank Handlowy w Warszawie, z kolei funkcję agenta dokumentacyjnego – PKO BP. (*kk*)

(http://media.tauron.pl/, 24.11.2015)

Spółka z Grupy Trans Polonia kupuje spółkę z Grupy ORLEN

Trans Polonia i PKN ORLEN porozumiały się w sprawie kupna/ sprzedaży 100% akcji spółki transportowej z Grupy ORLEN. Transakcja o wartości ok. 85 mln PLN obejmie wszystkie aktywa ORLEN Transport. Sprzedaż aktywów transportu samochodowego jest zgodna ze strategią PKN ORLEN, która zakłada koncentrowanie się na kluczowych z punktu widzenia Koncernu obszarach działalności: rozwoju segmentu Downstream, Detalu oraz Poszukiwań i Wydobycia. Spółka ORLEN Transport wejdzie z kolei do grupy, która świadczy specjalistyczne usługi w zakresie transportu i logistyki na terenie całej Europy. (*kk*)

(http://www.orlen.pl/, 16.11.2015)

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